

The analytical impact of sample storage on tritium analysis

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1. Introduction

Decommissioning of nuclear facilities will generate large volumes of wastes (e.g. concrete, asbestos, wood, desiccants, reactor metal work, graphite blocks and tiles, soft wastes etc). These may contain widely varying tritium activity concentrations and need to be characterized prior to waste sentencing. Careful consideration needs to be given to their sampling and storage if reliable analytical data are to be obtained. For this characterization, it is essential to take samples, store and transport before finally analyzing. There is a potential for cross contamination during each stage if care is not taken. This study aimed to determine ^3H emanating behaviour and degree of cross contamination under various storage conditions (e.g. ^3H form and origin, storage temperature and materials composition) and to consider the implication for sample storage and sampling strategy resulting in accurate ^3H analysis.

2. Methodology

A range of materials derived from various nuclear decommissioning sites, having different forms of ^3H , were used as tritium emanation source materials. A range of tritium free materials were used as potential receivers. Initial sample characterization and tritium extraction were performed using a Raddec Pyrolyser Trio System (Figure 1), which was designed specifically for quantitative extraction of tritium and ^{14}C from nuclear and environmental samples (Kim et al., 2008). All tritium measurements were performed using a 1220 liquid scintillation counter (Wallac QuantulusTM).

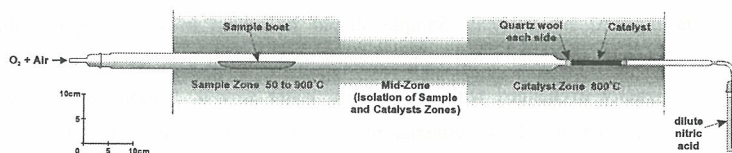


Figure 1. Schematic diagram of the Pyrolyser triple zone furnace

The magnitude of ^3H loss and hence cross contamination from concrete having different origin/form of ^3H was investigated. A known amount of active structural (~ 1 kBq) and bioshield (~ 6.6 kBq) concrete powder ($< 125\text{m}$, 1g) in a sealed polythene bag were placed into Kilner jar (635ml volume) modified with inlet and outlet ports which permitted the measurement of ^3H activity in the air of the container. Ten milliliters of RO water in an open and a closed vial were placed in each of the Kilner jars and the jars sealed and stored in a freezer (-18°C) and at room temperature (22°C). After 2 weeks, any ^3H activities measurable in the glass jars were determined.

3. Results and discussion

The magnitude of tritium emanation is dependent on the form of ^3H in the contaminated material and storage temperature. The most significant ^3H loss occurred at room temperature from structural concrete (non-irradiated concrete) where ^3H contamination arose from exposure of the concrete to HTO vapour (Table 1). For the

bioshield concrete which had been exposed to a neutron flux, ^3H loss is not significant. The greatest contamination transfer occurred to water in open vial from structural concrete at room temperature. Another non-negligible ^3H contamination transfer occurred to the cardboard which was used to support the open vial containing RO water. However, the proportions of ^3H activity transfers to water and cardboard from bioshield concrete were significantly less even at room temperature. For both structural and bioshield concrete, ^3H loss and cross-contamination is low for jars stored in the freezer. Significant quantities of ^3H contamination in water is due to the high isotopic exchange rate of tritium with water vapour where the reaction is significantly decreased as temperature is decreased (Numata et al., 1990; Munakata et al., 2003; IAEA, 2004; Furuichi et al., 2007). Tritium cross-contamination and its emanation behaviour depend significantly on the form of ^3H in the material and also on storage temperature.

Table 1. Tritium distribution after storage depend on ^3H form and storage conditions

| 3H contaminated into | Activity (Bq) of 3H (% of 3H) | | | |
|-------------------------------------|-------------------------------|-------------------|--------------------|-------------------|
| | Structural concrete | | Bioshield concrete | |
| | Room temp. | Freezer | Room temp. | Freezer |
| Air in the Kilner jar | 45.9 (4.1) | 4.4 (0.4) | 18.5 (0.3) | 2.7 (< 0.1) |
| Jar walls | 0.7 (0.1) | 0.1 (< 0.1) | 0.2 (< 0.1) | 0.1 (< 0.1) |
| Polythene bag (inside wash) | 5.2 (0.5) | 9.8 (0.9) | 8.0 (0.1) | 5.8 (0.1) |
| Polythene bag (outside wash) | 0.1 (< 0.1) | 0.1 (< 0.1) | 0.1 (< 0.1) | 0.1 (< 0.1) |
| RO water (open vial) | 377.1 (33.9) | 0.4 (< 0.1) | 282.4 (4.3) | 0.1 (< 0.1) |
| RO water (closed vial) | 0.2 (< 0.1) | 0.1 (< 0.1) | 0.2 (< 0.1) | 0.1 (< 0.1) |
| Out of scintillation vial | 0.1 (< 0.1) | 0.1 (< 0.1) | 0.1 (< 0.1) | 0.1 (< 0.1) |
| Cardboard a | 146.3 (13.2) | 17.7 (1.6) | 33.8 (0.5) | 5.2 (0.1) |
| 3H remaining in the concrete | 499 (45) | 1046 (94) | 6148 (94) | 6383 (97) |
| Total | 1075 (97) | 1078 (97) | 6491 (99) | 6397 (97) |
| Original 3H (Bq) in Concrete | 1112 (100) | 1112 (100) | 6577 (100) | 6577 (100) |

^a Cardboard support used with the open vial containing RO water

4. Conclusions

Careful consideration must be given to strategies for sample preservation in order to ensure that a representative sample is supplied for analysis. Sample should be sealed in vapour tight containers (glass or high density plastic is preferred) to avoid cross contamination and tritium loss, as the hydrogen from water or water-bearing samples is readily exchanged with atmospheric water vapour. Samples should be frozen to limit ^3H release and cross contamination. Low temperature can effectively reduce the tritium emanating rate therefore storing sample at freezer is the most secure and recommended way to preserve sample integrity. Finally, suspected low activity samples should be segregated from high activity samples to further reduce the possibility of cross contamination.

5. References

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